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Project Director: Dr. John W. Poston

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SPONSORED PROJECT TERMINATION/CLOSEOUT SHEET

Date 3/28/84

Project No. E-26-659 School/~~Lab~~ NE

Includes Subproject No.(s)

Project Director(s) Dr. J.W. Poston ~~GTRI~~ / GIT *

Sponsor Dept. of Energy, Oak Ridge, TN

Title Experimental Verification of Internal Dosimetry Calculations

Effective Completion Date: 1/31/82 (Performance) 1/31/82 (Reports)

* Changed to GTRI by Mod. A002 which is follow-on project E-26-621

Grant/Contract Closeout Actions Remaining:

- [X] None
- [ ] Final Invoice or Final Fiscal Report
- [ ] Closing Documents
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CaF_2 (Mn) suspended in organic liquids. Spurny and his colleagues[20] studied the effects on TL response of suspending LiF in water. The mechanism of the changes in response of TL materials in these liquids is not completely clear. The effects on TL response of suspending a TLD powder in an organic matrix (such as PAC) to mold "dosimetric organs" has not been investigated.

Proposed Research-Summary of Progress

During the past year the dosimetry research program has continued in the School of Nuclear Engineering and Health Physics at the Georgia Institute of Technology. The major objective of this program has been to provide research results upon which a useful internal dosimetry system could be based. The important application of this dosimetry system will be the experimental verification of internal dosimetry calculations such as those published by the MIRD Committee (1,2).

Specific research goals attained will be divided into several categories in order to discuss the progress in each area.

A. Preparation of Organ Models

One of the first challenges encountered during the last period was devising methods to produce a dosimetric organ for use in the experiments which matched as closely as possible the organs of the mathematical phantom. Initially, solid replicas of three organs were obtained from the Oak Ridge National Laboratory (ORNL). These replicas, which were fashioned from bakelite and coated with silicone, were used to produce organs for use in the original MR. ADAM.

Several techniques were investigated which could produce the organ molds for use in the experiment. The first technique studied was the use of labstone* and plaster of paris to produce a rigid mold into which the PAC

*Labstone is a material similar to plaster of paris which is used by the dental profession to prepare positive models of the teeth.

mixture could be poured and solidified. Procedures were formulated which were satisfactory for producing molds of small organs, e.g., kidneys. A detailed procedure for preparation of molds by this technique is given in Appendix I.

Molds produced by the above procedure were found to be too fragile for repeated use. Molds often chipped or completely shattered during routine use due to the stresses generated by the necessary clamping devices. A second technique for preparing molds was developed and the resulting molds will be used to produce the dosimetric organs for use in this study. This technique is described below.

Organs for use in the radiation dosimetry measurements were fabricated using the following procedures. A computer routine[21] was used to plot-out cross-sections of the phantom at 1 cm intervals along the vertical axis of the phantom. Organs, for which organ molds were required, were identified and the shapes were traced from the computer plots onto hardwood boards (1 cm thick). The shapes were cut out, assembled, and fabricated to create positive wooden replicas of the mathematical organs (see Figure 1). A second set of identical positive replicas was produced and sent to the University of Chicago for use in their studies. In this manner direct comparisons of the two experimental dosimetry techniques may be possible in the future.

The dosimetry technique suggested by Feher et al.[13] required a negative mold to produce a dosimeter which matches the organ shape. As mentioned above, several techniques were considered including plaster of paris - stone casting, vacuum plastic molding, and plaster bandage molding. The latter technique was selected because it was inexpensive, was relatively rapid, and the mold properties were superior to those obtained by other methods.

The procedure for forming negative organ molds is as follows. A hardwood mold was pre-treated with a thin film of silicone lubricant to provide swift

removal of the plaster mold. Several layers of the quicksetting plaster bandages* were mixed with water and wrapped around the pretreated mold. Bandages were applied to the mold until a layer approximately 2 cm thick was formed. The plaster "cast" was allowed to harden, at room temperature for a short time until it was sufficiently rigid to be cut. Then the cast was cut into two pieces for future removal of the organ mold. However, the mold was allowed to set-up completely for more than 18 hours, at room temperature, with the positive mold still inside to avoid shrinkage and assure structural integrity.

After removal of the positive organ mold the two halves of the plaster (negative) mold are rejoined and held together with additional strips of the plaster bandages (see Figure 1). These negative molds, produced by the above procedures, will be used to form the required volumetric dosimeters. The dosimeter itself will be formed from combinations of tissue equivalent materials and thermoluminescent powders. The intial material is a liquid which is introduced into the negative mold and is solidified rapidly (see Figure 1). This procedure will be discussed in a subsequent section of this progress report.

B. Preparation of Organ Dosimeters

Initially organ dosimeters were to be fabricated using a combination of paraffin, alkyl-chloride, and thermoluminescent powder (LiF). Samples of several diferent n-alkyl-chlorides were obtained from a commercial vendor. Organs molded from this material were extremely soft at room temperature and were extremely difficult to handle without deforming the organ shape. In

*Johnson and Johnson Products, Inc., New Brunswick, NY 08903

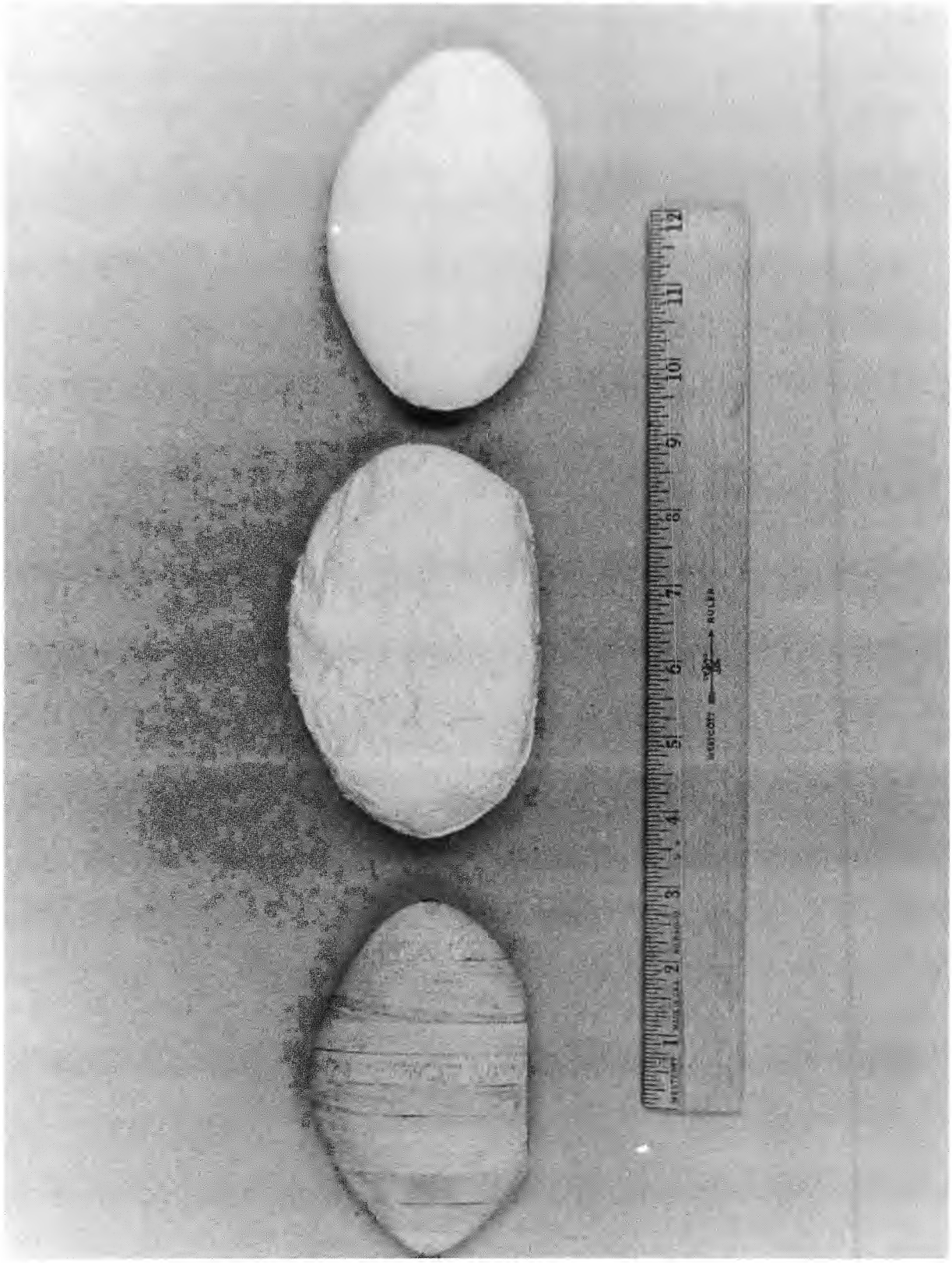

Figure 1. Photograph of wooden organ (left), plaster of paris mold (center), and PATE organ dosimeter for the kidney.

addition, if left at room temperature, the alkyl chloride "weeped" from the organ volume. No effort was made to quantify the loss of material from the organ dosimeters but it was a cause for concern and the alkyl-chloride was studied further.

Large samples of the alkyl chloride material were chemically refined to remove impurities in an attempt to produce a material which would not "weep" when used in the PAC material. These experiments indicated that the alkyl-chloride samples contained large quantities of confined water. In addition, it was revealed that samples contained an unacceptable amount of foriegn material. These studies indicated that the available alkyl-chloride compounds would not be acceptable for use in this research project unless extensive purification procedures were developed.

Studies of the PAC-paste material using the computer code TECALC [22] indicated that the photon interaction cross-sections did not match those of the tissue it was intended to represent. In addition, the density of the PAC-paste was determined to be 0.87 gm/cm^3 rather than the essentially 1 gm/cm^3 required to match tissue.

For the above reasons, it was decided to delay fabrication of organ dosimeters using the PAC mixture and to initiate a search for other acceptable substitute materials. Several substances were identified as possible substitutes but only one of these was currently available at a cost which allowed extensive use in the proposed research. This material was tetrachlorobenzene ($C_6H_2Cl_4$).

The organ dosimeter prepared by mixing paraffin, tetrachlorobenzene, and thermoluminescent powder was called the PATE dosimeter. This dosimeter was composed of 2% TLD-100, 11.2% $C_6H_2Cl_4$, and 86.8% paraffin. Table 1 presents a summary of the general characteristics of the organ dosimeter.

TABLE 1. General Characteristics of the PATE Organ Dosimeter

Composition by weight	
TLD-100	2%
Tetrachlorobenzene	11.2%
Paraffin	86.8%
Effective Atomic Number:	7.35
Number of Electrons Per Gram:	3.41×10^{23}
Average Atomic Weight:	11.87
Net Chemical Composition	
Hydrogen	13.8%
Carbon	78.6%
Lithium	0.5%
Fluorine	1.5%
Chlorine	5.7%

The PATE dosimeter is prepared according to the following procedure:

a. screen the TLD-100 powder through the 100 and 200 Tyler mesh screens to ensure uniformity of the powder.

b. grind the tetrachlorobenzene into a fine powder and pass it through the 200 Tyler mesh screen.

c. weigh out the proper amounts of the constituents of the PATE organ dosimeter.

d. combine the paraffin and tetrachlorobenzene and melt at a temperature of 75-80^{o}C while stirring the mixture.

e. when melting is complete, pour the mixture through the 200 Tyler mesh screen and collect the resultant liquid.

f. add the weighed amount of TLD-100 powder to the mixture and agitate to insure uniform mixing.

g. allow the mixture to cool, while stirring, to about 50^{o}C, then pour the mixture into the mold.

h. allow the mixture to cool and solidify completely before removing from the mold.

The mold itself is prepared for use by lining the inside of the mold with aluminum foil. The foil facilitates removal of the organ dosimeter from the mold. Other coating materials, such as silicone, were transferred from the mold to the PATE material upon cooling. The contamination of the organ dosimeter by this material led to the use of aluminum foil to line the inside of the molds. To expedite rapid cooling of the PATE mixture the mold is cooled in a deep freezer and, in some cases, the pouring is carried out in the freezer. In some cases additional PATE must be added to the mold because of shrinkage due to cooling.

The above procedure produces an organ dosimeter with qualities much superior to those obtained with the PAC mixture. The organ has a pure white color and the consistency of the material is much harder than the previous material. Tetrachlorobenzene has a higher melting point and there is no evidence of weeping or other mechanisms which may lead to a loss of material. Photon attenuation cross-sections determined with TECALC [27] and the density of the PATE material match those of tissue more closely.

C. Recovery of TLD from the PATE Organ Dosimeter

Techniques for the recovery of the thermoluminescent powder from the organ dosimeter were studied extensively during the past year. These studies were the result of a change from alkyl-chloride to tetrachlorobenzene as a major constituent of the dosimeter. The current procedure used for recovery is as follows:

a. place the organ dosimeter in a large beaker and add carbon tetrachloride (CCl_4) in a 1:2 ratio by volume.

b. heat the mixture while keeping the temperature at or below $80^{\circ}C$.

c. once the PATE mixture has dissolved, pour the entire mixture through the 200 Tyler mesh screen and wash the beaker with extra CCl_4 until all TLD-100 crystals are removed.

d. wash the screen with more CCl_4 until the TLD crystals appear to be completely free of paraffin.

e. allow the TLD crystals to air-dry while on the screen.

f. once the crystals are dry, pass the recovered crystals through the 100 and 200 Tyler mesh screens to remove any collected impurities.

g. before evaluating the TLD crystals, preanneal the crystals at $100^{\circ}C$ for 10 minutes.

h. read the TLD crystals on a thermoluminescent dosimeter reader following manufacturer's recommended procedures for heating rate, maximum temperature, etc.

Recovery of TLD crystals by the above procedure has been quite good with an average recovery rate of about 96%.

D. Distribution of the TLD in the PATE Organ Dosimeter

A study was initiated to determine the ability of the molding technique (described in B. above) to produce an organ dosimeter which had a uniform distribution of the TLD powder. A cylindrical plaster-bandage mold was fabricated for use in this study. The inside of the cylinder had a diameter of 5.3 cm and a height of 30 cm.

A 500 g sample of the PATE mixture was prepared following the procedures described above. The PATE mixture contained 10.0 g of TLD-100, 55.85 g of tetrachlorobenzene, and 434.15 g of paraffin. The mold was kept in the freezer until the PATE mixture was ready for pouring. After filling the mold with liquid PATE, the mold was replaced in the freezer in a vertical position.

After cooling, the solidified PATE cylinder was removed from the plaster-bandage mold and cut into 13 slices. The thickness and mass of each slice was determined before attempting to recover the TLD-100 powder. The data obtained in this study are summarized in Table 2. Note that the PATE mixture prepared for this experiment was not sufficient to fill the mold completely. This fact is reflected in column 3 of the table. Slice number one corresponds to the bottom of the cylinder whereas slice number thirteen is the upper portion of the cylinder. Note the extremely uneven distribution of the TLD-100 powder in the mold. In addition, note that the TLD recovery in this experiment was only approximately 81%. Further investigation of the glassware used in the experiment revealed that 1.41885 g of material had

remained in the beaker and the funnel. Subsequent refinement of the recovery procedure (discussed previously) indicates that the recovery may ultimately approach 100%.

The data presented here are representative of several of the distribution studies performed to date. The skewed distribution indicates that other procedures must be developed in order to ensure a uniform distribution of the TLD powder in the organ dosimeter. At the present time, organ molding techniques are being investigated which utilize a layering process to overcome these problems. Preliminary results are given below which indicate that this technique should be successful. Inspection of the complete PATE molded organs has shown good joining of the layers. In fact, once the mold has solidified the individual layers are not identifiable to the naked eye. No boundaries are visible and the layered organ dosimeters exhibit similar physical characteristics to those molded at one pouring.

E. Modified Molding Techniques

As mentioned above, modified molding techniques have been developed to overcome the TLD setting problem. In this modified procedure the paraffin and tetrchlorobenzene are combined, melted and solidified before the organ molding procedure is begun. The procedure is as follows:

a. melt paraffin and $C_6H_2Cl_4$ at a temperture of about 80-100°C.

b. pour this mixture through the 200 Tyler mesh screen.

c. allow the mixture to solidify in a shallow stainless steel pan.

d. when organ dosimeters are to be formed, break the paraffin-tetrachlorobenzene material into chunks.

e. place about 100 g of these chunks in a beaker and remelt at about 80-100°C.

TABLE 2. Results of TLD Distribution Study

Slice Number	Height (cm.)	Cumulative Height (cm.)	Weight (g)	TLD-100 Weight (g)	TLD Concentration (%)
1	2.042	2.042	45.05492	7.51645	16.6829
2	1.431	3.473	31.57830	0.29480	0.9336
3	1.552	5.025	39.23465	0.13410	0.3917
4	1.137	6.162	25.08504	0.03795	0.1513
5	1.467	7.629	32.37079	0.03720	0.1149
6	1.082	8.711	23.87346	0.02165	0.0907
7	1.567	10.278	34.56629	0.02178	0.0360
8	1.532	11.810	33.79788	0.02650	0.0783
9	1.972	13.782	43.51380	0.02220	0.0510
10	1.562	15.644	34.46477	0.01388	0.0402
11	1.922	17.566	42.41088	0.01012	0.0239
12	1.455	19.021	32.09712	0.00005	0.0002
13	1.617	20.638	35.66424	0.01068	0.0299

Total weight of TLD recovered 8.14735 g

f. screen the TLD-100 crystals through the 100 and 200 Tyler mesh screen.

g. weigh out proper amount of the TLD-100 cyrstals for use.

h. pour a portion of the melted mixture into a 50 ml beaker and add the correct amount of TLD powder. Stir the mixture well and pour the liquid PATE into another beaker for future use.

i. pour another 50 ml of the melted mixture into the same beaker and add the required amount of TLD powder. Pour this liquid PATE into the organ mold and allow 10-15 minutes for solidification.

j. repeat step i, allowing for solidification, until the mold is completely filled.

The distribution study described in D. above was repeated using the above described procedure. The right circular cylinder of PATE was cut into 17 slices. Each slice was measured, dissolved and the TLD crystals recovered according to established procedures. The data from this experiment are shown in Table 3. The mean concentration in the 17 slices is 1.086 $\pm$ 0.470 g of TLD powder. However, close inspection of the table indicates a problem with top four slices (slices 14-17). The high concentration in slice 14 is thought to be due to the lack of aherence to the established procedure. That is, the PATE layer was not allowed to solidify completely before subsequent layers were poured. The mean concentration in slices 1-13 was found to be 1.120 $\pm$ 0.216 g of TLD powder. This variation in the TLD distribution ($\pm$ 20%) is considered acceptable for use in the present study. However, further refinements are being studied at this time which are expected to reduce the present variations.

F. Effect of the PATE Mixture on the TLD Response

Before the PATE mixture can be used successfully in this dosimetry research the response characteristics of the TLD powder must be investigated. Preliminary investigations on the PAC material (reported last year) indicated no significant changes in response. However, the subsequent change from alkyl chloride to tetrachlorobenzene has necessitated further study of the TLD response characteristics. The results of these studies are not available for inclusion in this progress report. Nevertheless, preliminary results indicate that any change in response may actually be due to incomplete removal of the paraffin from the crystals. The modified recovery procedure (discussed in C. above) was designed to insure the recovery of clean crystals.

G. Investigation of Chemical Dosimetry Systems

A preliminary literature survey, reported in the last progress report, indicated that the ferrous sulphate-benzoic acid-xylenol orange dosimetry system (FBX) had characteristics suitable for use in this research. The FBX system consists of 0.2 mM ferrous ammonium sulphate, 5.0 mM benzoic acid, and 0.2 mM xylenol orange in 0.05 N sulphuric acid. The dosimeter could be used for dosimetry of X, gamma, and neutron radiation in the dose range of 0.1 - 3000 rad. The dosimetric solutions could be stored for about 2 weeks before irradiation and up to 3 days after irradiation without any significant error in dose estimations (22).

Further investigation of the FBX system indicated that this system was not suitable for use in this research. Although the dose range reported in the literature was 0.1 - 3000 rads the accuracy at 100 mrad was $\pm$ 50 mrad. The chemical procedures and equipment required to obtain even this level of accuracy were also prohibitive. Subsequent publications by the same authors

TABLE 3. Results of TLD Distribution Study Using Modified Molding Techniques

Slice Number	Height (cm)	Cumulative Height (cm)	Total Weight (g)	TLD-100 Weight (g)	TLD Concentration (g)
1	0.68	0.68	14.95150	0.19273	1.289
2	0.88	1.56	19.37895	0.24385	1.258
3	1.42	2.98	31.28255	0.43510	1.390
4	1.22	4.20	26.83448	0.35420	1.320
5	1.72	5.92	38.01695	0.34540	0.909
6	1.23	7.15	27.21747	0.21915	0.805
7	1.03	8.18	22.68863	0.24100	1.062
8	1.42	9.60	31.38180	0.38513	1.227
9	1.78	11.38	39.23075	0.43495	1.109
10	1.25	12.63	27.58180	0.20190	0.732
11	1.12	13.75	24.61440	0.24365	0.990
12	1.31	15.06	28.8653	0.40120	1.390
13	1.10	16.16	24.25600	0.26130	1.077
14	1.77	17.93	38.95190	0.93115	2.391
15	1.37	19.30	30.30130	0.27295	0.901
16	1.39	20.69	30.6400	0.11150	0.364
17	2.98	23.67	65.84770	0.16225	0.246

(23) conculded that use of a spectrophotometer with a 10 cm cell length probably had a sensitive limit in the range of 1-4 rads rather than 100 mrad reported earlier. This technique was not investigated further due to the stated accuracy in the dose range of interest.

Another system which was investigated used a liquid photographic emulsion (24). This dosimeter was designed to be useful in the 0.5 - 6R range. The average deviation in response was 2.5% or less for paired samples. The concentrations used were approximately tissue equivalent and the variation in response from 16 keV to 1.33 MeV was less than 50%.

Discussions with one of the developers of this system revealed that the system, as described in the literature, did not function as well as the initial results indicated. Many problems were encountered in the routine use of this dosimetry system and the idea was abandoned in favor of suspending TLD powder in a silicon liquid media. However, this latter system also was never fully developed (25).

A broad-range chemical dosimetry system described by Klein (26) was also investigated. This system used saturated aqueous solutions of benzene and could be used to measure accurately doses as low as 10 R. The evaluation at this exposure level required the analysis of the radiolysis product phenol in the 10^{-8} molar concentration range. This system was not considered further due to the capabilities available in our laboratory and the necessity of reducing the sensitive exposure range by two orders of magnitude.

H. Calculations of Absorbed Doses by Monte Carlo Techniques

No final calculations have been performed with which experimental comparison can be made. Computer codes to be used in this research have previously been installed on the Georgia Tech computer. These codes have been debugged and computer results have been compared to test cases supplied with

the code. This phase of the research can be completed within one or two weeks once the exact tissue-equivalent compositions to be used in the experiments have been established. A significant effort has been expended to understand the Monte Carlo code and the methods to be used to modify the code before producing the final computer data. This effort is intended to allow recalculation of internal exposure situations which match as closely as possible the experimental situation.

The computer code TECALC[27] has been used extensively to design tissue-, bone-, and lung-equivalent materials for use in the MR. ADAM phantom as well as the selection of materials to replace the PAC-TLD paste.

Research Goals for the Renewal Period

Current progress in this research, coupled with the remaining four months of the project, indicate that the research goals will be attained as scheduled. However, there are several areas which remain unresolved and for which a renewal of this research effort is requested. These areas are discussed below.

A. Consideration of Additional Organs

Organ molds have been fabricated only for a small number of organs which were selected at the inception of this research. It is proposed that the initial dosimetry investigation be extended to include many additional organs. Further, it is proposed that complex organs and organ systems be studied for inclusion in the research.

It is proposed that studies be undertaken to investigate the fabrication of organ dosimeters for organs which comprise the more complex organ systems in the body. For example, the lungs have a density of 0.3 g/cm^3 whereas the present organ dosimeters are designed to simulate tissues with density of 1.0 g/cm^3. In previous studies the lungs have been simulated effectively in order to produce the proper scattering and transport medium. However, no attempts